



Assembly of quantum dots in polymer solar cells driven by orientational switching of mesogens under electric field

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Abstract

In-situ synthesis and rapid assembly of CdS quantum dots (QDs) in main-chain liquid-crystalline polymer poly(2,5-bis(3-alkylthio phen-2-yl)thieno[3,2-b]thiophene) (PBDTTT-C12) and side-chain liquid-crystalline polymer poly[4,8-bis(2-ethylhexyloxy)benzo[1,2-b:4,5-b']dithiophene-alt-3,6-bis(thiophen-5-yl)2,5-dihydropyrrolo[3,4-c]pyrrole-1,4-dione] (PBDTDPPcbp) was formed. This was driven by the organization of liquid-crystals (LC) at 150 °C, due to mesotropic state of mesogens. Further under the applied electric field (V_{app}), the orientation of QDs could be regulated along with the highly crystalline nanofibers to form the separated organic and inorganic orderly channels, which was benefit for the electron transportation and collection. The inverted polymer solar cells (i-PSCs) devices with ZnO modified by PBDTTT-C12@CdS and PBDTDPPcbp@CdS interfacial layers (ILs) after rinsing polymers with chloroform as electron transport layers (ETLs) and poly(3-hexylthiophene-2,5-diyl):[6,6]-phenyl-C₆₁-butyric acid methyl ester (P3HT:PC₆₁BM) as active layer showed superior photovoltaic performance with average power conversion efficiency (PCE) of 3.5% and 3.9%, respectively. With application of suitable V_{app} , the more orderly CdS QDs interfacial nanostructures were in favor of the higher electron mobility and better interfacial contact between cathode and active layer, consequently enhanced the devices performance.

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1. Introduction

The optoelectronic properties of the photoactive layer determine the maximum photocurrent generation and efficiency of the polymer solar cells (PSCs) (Park et al., 2009; Seo et al., 2011; H.Y. Chen et al., 2009). The interfacial layers sandwiching between the photoactive layer and corre-

sponding electrodes are of equal importance as they could provide efficient charge carrier extraction toward the electrodes avoiding hereby losses such as charge carrier recombination and exciton quenching at these interfaces.

As the electron transport layer (ETL) of the inverted polymer solar cells (i-PSCs), zinc oxide (ZnO) is the most common candidate due to its easy synthesis, relative high electron mobility, together with suitable energy level for electron extraction from active layer (Dkhil et al., 2014; Lee et al., 2014; Cui et al., 2013). To modify the surface defect of ZnO and further increase the electron transport ability, it is wise to select cadmium sulfide quantum dots (CdS QDs) as modifiable candidate, which can decrease

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the charge recombination contributing to better charge extraction efficiency (Cui et al., 2013). To further reduce the energy barrier between the cathode and active layer and form well energy level of device, interfacial dipole layers could be applied via surface modification of ETL such as metal oxide or inorganic nanocrystals (Zhang et al., 2014; Yang et al., 2012). The insertion of aligned molecular dipole moments at the organic/inorganic (O/I) interface with a favorable bulk nanostructure will dwindle the energy level difference between the organic polymer and inorganic nanocrystals, consequently strongly affecting the performance of devices. Furthermore, it is possible that the interfacial properties could be regulated through response to various external stimuli, such as the electric field. Tajima et al. had reported that electric field-induced dipole switching at the donor/acceptor interface in organic solar cells (Zhong et al., 2013).

In general, with the help of liquid-crystalline (LC) alignment, well-organized nanostructure of liquid-crystalline copolymers could be fabricated with excellent electrical properties for preparing nanomaterials (Yu et al., 2011). It had also been displayed the orientation of liquid-crystalline block copolymers with hydrogen-bonded side-chains could be regulated by alternating the current of electric field (Chao et al., 2004). In this work, we showed that the microstructures of liquid-crystalline copolymers can be rapidly aligned in the applied electric field (V_{app}), because of the incorporation of mesogenic groups with marked dipolar properties. A mechanism related to LC alignment is proposed to account for the fast orientational switching of the liquid mesogens (Shi et al., 2013). I-PSCs devices with the in-situ synthesis of CdS QDs in main-chain liquid-crystalline polymer poly(2,5-bis(3-alkylthiophen-2-yl)thieno[3,2-b]thiophene) (PBDTTT-C12) (McCulloch et al., 2006) and side-chain liquid-crystalline polymer poly[4,8-bis(2-ethylhexyloxy)benzo[1,2-b:4,5-b']dithiophene-alt-3,6-bis(thiophen-5-yl)2,5-dihydropyrrolo[3,4-c]pyrrole-1,4-dione] (PCBTDPpCbP) (Han et al., 2013) respectively as interlayers (ILs) on the ZnO surface were fabricated. The ILs with different composition and oriented nanomorphologies by applying V_{app} ranging from 0 V/mm to 600 V/mm were also studied.

2. Results and discussion

It is well-known the orientation of liquid-crystalline polymers can be regulated by alternating the current of electric field (Chao et al., 2004), attributing to their long-range ordered fluidity, molecular motion and alignment change by external V_{app} . Herein, main-chain liquid-crystalline PBDTTT-C12 and side-chain liquid-crystalline PCBTDPpCbP were used to drive the in-situ growth and assembly of CdS, induced by the organization of LC mesogens (Choi et al., 2004; L. Chen et al., 2009) in these polymers upon thermal annealing and further applied V_{app} (in Scheme 1). The PCBTDPpCbP in this work was synthesized via Suzuki–Miyaura polymerization reactions and the

detailed route was provided in Supporting Information (as shown in Scheme S1). The PBDTTT-C12 and PCBTDPpCbP were chosen because of their LC mesogens in different locations, main-chain backbone and side-chain respectively, and the corresponding various assembly property driven by applying considerable V_{app} intensity. The CdS precursor preferred binding to the sulfur atoms from PBDTTT-C12 backbone (LC mesogens) and PCBTDPpCbP conjugated backbone, which was consistent with the previous report (Liao et al., 2009). This suggested that there might be stronger assembly of CdS QDs driven by the LC mesogens for PBDTTT-C12@CdS. Upon thermal annealing at 150 °C, the CdS QDs were synthesized and the morphologies of embed CdS in PBDTTT-C12@CdS and surface dispersed CdS in PCBTDPpCbP@CdS were formed upon further applied V_{app} with intensity of 200 V/mm. After rinsing polymers with chloroform (CHCl₃), r-PBDTTT-C12@CdS (V_{app} = 200 V/mm) and r-PCBTDPpCbP@CdS (V_{app} = 200 V/mm) were applied in PSCs as the ILs. “r-” denotes as PBDTTT-C12@CdS (V_{app} = 200 V/mm) and PCBTDPpCbP@CdS (V_{app} = 200 V/mm) rinsed by CHCl₃.

The ultraviolet–visible (UV–Vis) absorption spectra in Fig. 1 are conducted to confirm the in-situ formation of CdS QDs into the liquid-crystalline polymers, resulting from the larger absorption of PBDTTT-C12@CdS and PCBTDPpCbP@CdS below 500 nm, comparing with pure PBDTTT-C12 and PCBTDPpCbP. This characteristic is attributed to the absorption of CdS QDs. Taking into account that the UV–Vis absorption properties of PBDTTT-C12@CdS and PCBTDPpCbP@CdS ILs have the negative effect on the device performance, further treating the ILs surface with CHCl₃ has been carried out. The absorption peaks at 400–600 nm and 300–750 nm originated from the main chain of PBDTTT-C12 and PCBTDPpCbP reduce mostly comparing with the as-cast sample without CHCl₃ rinse, indicating liquid-crystalline polymers could be rinsed with no effect on the CdS.

As shown in Fig. S1, the X-ray photoelectron spectroscopy (XPS) spectra of S 2p and Cd 3d in PBDTTT-C12@CdS and PCBTDPpCbP@CdS have illustrated the successful in-situ growth of CdS QDs in PBDTTT-C12 and PCBTDPpCbP polymer chains. Compared with the two Cd peaks for PCBTDPpCbP@CdS at 411.7 and 404.96 eV, the corresponding Cd peaks for PBDTTT-C12@CdS shift to 411.36 and 404.58 eV with lower energy values (Shi et al., 2014), indicating the stronger interaction between CdS and PBDTTT-C12. Moreover, the intermediate bonding energy of S 2p peaks for PBDTTT-C12@CdS and PCBTDPpCbP@CdS can also be observed, explaining the possible orderly assembly of CdS QDs in the polymer chains, which will be further confirmed by transmission electron microscopy (TEM) measurement below.

Due to PBDTTT-C12 and PCBTDPpCbP with LC mesogens in different locations, main-chain backbone and side-chain respectively, the corresponding driven assembly of CdS by applying V_{app} may exist difference. Observed

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